Residues in apples and sweet cherries after methyl bromide fumigation†

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Abstract: Methyl bromide fumigations are used to treat apples, *Malus domestica* Borkh, and sweet cherries, *Prunus avium* (L), before export to Japan. In order to expand existing markets, additional cultivars are being prepared for export to Japan. As part of the approval process, residue analyses must be conducted and residues must be at acceptable levels. Five apple cultivars ('Braeburn,' 'Fuji,' 'Gala,' 'Jonagold,' and 'Granny Smith') were fumigated at 40 g m^{-3} for 2 h at 10 °C, and six sweet cherry cultivars ('Brooks,' 'Garnet,' 'Lapin,' 'Rainier,' 'Sweetheart,' and 'Tulare') were fumigated for 2 h with 64 g m^{-3} at 6 °C, 48 g m^{-3} at 12 °C, 40 g m^{-3} at 17 °C, and 32 g m^{-3} at 22 °C. Three replicates of fruit from each fumigation were analyzed for methyl bromide and bromide ion residues periodically with time. Methyl bromide residues for both apples and cherries were the highest immediately after fumigation, but rapidly declined so that only 'Braeburn' had residues >8 µg kg^{-1} after 13 days and, except for 'Lapin,' all cherries were <1 µg kg^{-1} after seven days. Average bromide ion residues were between 3.3 and 4.9 mg kg^{-1} among apple cultivars, and between 3.7 and 8.0 µg kg^{-1} among cherry cultivars.
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1 INTRODUCTION

Some countries, such as Japan, require that US-produced apples, *Malus domestica* Borkh, and sweet cherries, *Prunus avium* (L), must be fumigated with methyl bromide for codling moth, *Cydia pomonella* L (Lepidoptera: Tortricidae), an internal-feeding quarantine pest.[1–3] Apples are fumigated with 56 g m^{-3} for 2 h at 10 °C after 55 days of cold storage at 2.2 °C, and sweet cherries are fumigated with 64 g m^{-3} for 2 h at 6 to 12 °C; 48 g m^{-3} for 2 h at 12 to 17 °C; 40 g m^{-3} for 2 h at 17 to 22 °C; or 32 g m^{-3} for 2 h at 22 °C and above. The difference in storage and treatment temperatures for apples provides for treatment efficacy with reduced phytotoxic effects. Variation in the treatment schedules allows for flexibility in the timing of fumigation, whether the fruit are directly treated from the orchard or are first put in cold storage. Determination of methyl bromide and bromide ion residues from the fumigation regimes is required by Japan for approval of the treatment schedules. Bromide residues from quarantine treatments have been examined for Hawaiian fruits and vegetables,[4,5] Australian fruits and vegetables,[6] nectarines,[7,8] sweet cherries,[7,9] apples,[10] and other deciduous temperate fruits.[7]

Typically, the methyl bromide is rapidly expelled from the free airspace in the chamber within the first 30 min of aeration, and this is followed by a more gradual release from the fruit.[10] Factors influencing release from a commodity are its oil and moisture content, fumigation temperature, and fumigant dosage.[11] Ultimately, methyl bromide breaks down to bromide ion, which is relatively innocuous.[6]

Herein, we report the methyl bromide and bromide ion residues resulting from various fumigation schedules for five cultivars of apples and six cultivars of sweet cherries.

2 MATERIALS AND METHODS

2.1 Test fruits

Tests were conducted with mature unwaxed Fancy or Extra Fancy size 113 (113 fruits per carton) fruits of cultivars 'Braeburn,' 'Fuji,' 'Gala,' 'Jonagold,' and 'Granny Smith.' These apples were obtained from commercial fruit packing and marketing concerns in

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Washington state. Mature, field-run (no size selection or culling) 'Lapin' and 'Sweetheart' sweet cherries were obtained from commercial orchards in Washington state; 'Brooks,' 'Garnet,' 'Rainier,' and 'Tulare' sweet cherries were obtained from central California. All fruits were produced following established pest-control practices in the orchard and were later stored (at $\simeq 2\,°C$) and treated at the USDA-ARS Laboratories in Yakima and Wapato, Washington.

2.2 Treatment and sampling

All fumigations were conducted in 28.3-litre fibreglass chambers (Labconco Vacuum Desiccators, Model 55300, Labconco Corp, Kansas City, MO) each equipped with a circulating fan which operated continuously throughout the exposure period. The chambers were in a walk-in controlled-environment room. The temperatures in the chambers were controlled by the temperature of the room. Fruits were held in each chamber within a perforated metal basket designed to allow flow of the fumigant and to produce a 50% load factor (fruit volume: chamber volume) when filled. Temperatures were monitored during fumigations using a computer-controlled temperature recorder equipped with thermisters; for each chamber, a thermister was inserted into one fruit placed in the center of the load. Concentrations of methyl bromide in the air space in the chamber were monitored by gas chromatography.

Fumigations of the 'Brooks,' 'Garnet,' 'Rainier,' and 'Tulare' cultivars were conducted at an older facility in Yakima, WA, whereas fumigations of all the apple and remaining cherry cultivars were done at a newly built laboratory near Wapato, WA. Each apple cultivar was treated with $56\,g\,m^{-3}$ for 2-h at $10\,°C$, followed by a 2-h aeration period; the fruits subsequently were placed in export cartons (as specified by MAFF-Japan requirements) and stored at $\simeq 2\,°C$ for up to 13 days. The methyl bromide treatment regimes used for cherries were 2-h fumigations with: $64\,g\,m^{-3}$ at $6\,°C$, $48\,g\,m^{-3}$ at $12\,°C$, $40\,g\,m^{-3}$ at $17\,°C$, and $32\,g\,m^{-3}$ at $22\,°C$. After fumigation and a 2-h aeration period, cherries were placed in commercial shipping cartons with polyethylene liners at simulated shipping temperatures ($\simeq 2.5\,°C$) for up to seven days. Periodically (ie for apples, after 2 h and 1, 3, 5, 7, 9, 11, and 13 days post-treatment; for cherries, after 5 h and 1, 2, 3, 5, and 7 days post-treatment), $\simeq 500\,g$ of fumigated material was sampled, frozen, and sent to the USDA-ARS Horticultural Crops Research Laboratory in Fresno, California, for residue analysis. The apple samples were transported in a freeze unit and the cherry samples were shipped with dry ice.

2.3 Residue analysis

To determine methyl bromide residues, three diced fruit samples (50 g for apples, 100 g for cherries) for each sample interval were taken at random from each cultivar replicate for each treatment. Equal amounts of the same fruits were taken as controls. Following minor modifications of the method described by King *et al*[12] and Hartsell *et al.*[13] methyl bromide residues were determined by assaying the headspace with a gas chromatograph. The limit of detection using this method with matrix present was $0.001\,\mu g\,g^{-1}$ ($=0.001\,ppm = 1\,ppb$).

Bromide ion residues were determined using an X-ray fluorescence spectrophotometer (Spectrace 431, Tracor Northern, Middleton, WI) according to the method described by Getzendaner *et al.*[14] and modified by Harvey *et al.*[15] Three diced fruit samples (50–100 g) were taken randomly from each cultivar replicate for each treatment and held for three days at $\simeq 2.5\,°C$ before analysis. The inorganic bromide was extracted using water as a solvent. After extraneous organic material had been eliminated by ashing, the bromide ion was oxidized with sodium hypochlorite to bromine and subsequently titrated with a standard sodium thiosulfate solution. The limit of detection was $2\,\mu g\,g^{-1}$ ($=2\,ppm$).

2.4 Data analysis

Percentage sorption was calculated by dividing the difference between the first and last measured concentrations during fumigation by the first concentration, then multiplying by 100%. Statistical analyses of the data were conducted by using SAS.[16] Methyl bromide residue data were transformed to their natural logarithm to fit the data to the Tebbets model,[7] then regressed using PROC GLM. Pair-wise comparisons between the regression coefficients were made using the equation,

$$t = (b_1 - b_2)/S_{b1-b2} \tag{1}$$

where t is the Student t value, b_1 and b_2 are the

Table 1. Mean methyl bromide residues in apple cultivars after methyl bromide fumigations at $56\,g\,m^{-3}$ for 2 h at $10\,°C$ followed by 2 h aeration and storage in export cartons at $\simeq 2\,°C$ for 13 days. Controls had $<1\,\mu g\,kg^{-1}$ methyl bromide residue

	Methyl bromide residues ($\mu g\,kg^{-1}$) ($\pm SD$)[a]							
Cultivar	*2h*	*1 day*	*3 days*	*5 days*	*7 days*	*9 days*	*11 days*	*13 days*
'Braeburn'	7800 (±700)	4300 (±500)	950 (±80)	400 (±80)	180 (±60)	60 (±20)	16 (±2)	8.0 (±5)
'Gala'	14000 (±2000)	3800 (±200)	490 (±5.0)	27 (±6)	5.0 (±0)	6.0 (±3)	3.0 (±0)	<1
'Fuji'	10000 (±1000)	4300 (±600)	870 (±10)	180 (±30)	51 (±4)	17 (±4)	3.0 (±1)	1.0 (±1)
'Granny Smith'	10000 (±800)	3600 (±200)	690 (±60)	210 (±60)	50 (±20)	14 (±3)	3.0 (±1)	2.0 (±0)
'Jonagold'	8000 (±3000)	2700 (±1000)	420 (±50)	30 (±10)	6.0 (±4)	2.0 (±0)	<1	<1

[a] $n = 3$.

Table 2. Mean bromide ion residues in apple and sweet cherry cultivars after methyl bromide fumigation. Controls for apple cultivars had $<2\,mg\,kg^{-1}$ bromide ion residue and controls for cherry cultivars had $<1\,mg\,kg^{-1}$ bromide ion residue.

		Bromide ion residue[a] *(mg kg^{-1}) (± SD)*[b] *by dose (mg m^{-3})*				
Fruit	*Cultivar*	*32*	*40*	*48*	*56*	*64*
Apple	'Braeburn'				3.8 (±0.3)	
	'Gala'				3.6 (±0.5)	
	'Fuji'				3.3 (±0.3)	
	'Granny Smith'				4.1 (±1.0)	
	'Jonagold'				4.9 (±0.5)	
Cherry	'Brooks'	6.2 (±0.8)abc	5.5 (±0.5)abcd	6.0 (±0.5)abc		5.1 (±0.7)
	'Garnet'	4.4 (±0.3)ad	7.1 (±0.3)aef	6.4 (±0.6)de		6.2 (±0.4)ab
	'Lapin'	4.5 (±0.2)be	4.3 (±0.4)begh	3.7 (±0.8)adfg		5.7 (±0.4)c
	'Rainier'	8.0 (±0.9)defg	7.6 (±0.2)cgi	6.4 (±0.6)fh		6.7 (±0.8)d
	'Sweetheart'	4.3 (±0.3)cf	4.1 (±013)dfij	4.3 (±0.3)bhi		4.2 (±0.2)acde
	'Tulare'	5.2 (±0.6)g	5.6 (±0.2)fhj	6.0 (±0.1)egi		5.2 (±0.2)be

[a] Values in columns followed by the same letter significantly different ($P < 0.05$) as determined by Student's *t* test.
[b] $n = 3$.

regression coefficients (slopes) and s_{b1-b2} is the standard error of difference between the two regression coefficients.[17] Equation (1) also was used to determine significant differences between average inorganic residue measurements of cultivars treated with the same fumigant dose.

3 RESULTS AND DISCUSSION

3.1 Apples

Because methyl bromide is absorbed by the fruit tissue, washing will not reduce the residues. However, the fruit naturally expels the methyl bromide and the residues decrease with time. In our studies, the methyl bromide residues for the apple cultivars declined from a high of $16.1\,mg\,kg^{-1}$ ('Gala') 2 h after aeration to $<0.003\,mg\,kg^{-1}$ (for all except 'Braeburn') after 13 days (Table 1). The cultivar with the highest initial residues (2 h after aeration) was 'Gala,' whereas the lowest was 'Braeburn.' The longest-lasting residues were also with 'Braeburn.' In comparison, nectarines (treated with $48\,g\,m^{-3}$ for 2 h at $\geq 21\,°C$ in field bins) have been found to have methyl bromide concentration $<10\,\mu g\,kg^{-1}$ after two days (for 'Fantasia') and

Table 3. Mean methyl bromide residues in sweet cherry cultivars after three methyl bromide fumigations at four different doses for 2 h followed by 2 h aeration and storage in commercial shipping cartons with polyethylene liners at $\simeq 2.5\,°C$ for seven days

				Methyl bromide residues ($\mu g\,kg^{-1}$) (±SD)[a] *after treatment*					
Dose (g m^{-3})	*Cultivar*	*Temp (°C)*	*Sorption (%)*	*5 h*	*1 day*	*2 days*	*3 days*	*5 days*	*7 days*
32	'Brooks'	22	54.8	700 (±300)	7.0 (±1)	4.0 (±1)	3.0 (±1)	<1	<1
32	'Garnet'	22	55.7	1400 (±300)	<1	<1	<1	<1	<1
32	'Lapin'	22	56.9	29 (±9)	7.0 (±0)	4.0 (±1)	3.0 (±0)	2.0 (±1)	1.0 (±0)
32	'Rainier'	22	49.8	880 (±90)	<1	<1	<1	<1	<1
32	'Sweetheart'	22	52.6	180 (±50)	14 (±7)	4.0 (±3)	<1	<1	<1
32	'Tulare'	22	57.9	600 (±80)	6.0 (±1)	4.0 (±2)	4.0 (±1)	<1	<1
40	'Brooks'	17	53.9	900 (±500)	7.0 (±2)	6.0 (±1)	4.0 (±1)	<1	<1
40	'Garnet'	17	55.8	2200 (±600)	200 (±200)	2.0 (±2)	<1	<1	<1
40	'Lapin'	17	58.1	40 (±10)	6.0 (±1)	4.0 (±0)	4.0 (±1)	2.0 (±0)	2.0 (±1)
40	'Rainier'	17	55.7	1700 (±100)	6.0 (±5)	<1	<1	<1	<1
40	'Sweetheart'	17	55.5	290 (±30)	30 (±8)	6.0 (±4)	<1	<1	<1
40	'Tulare'	17	56.7	1500 (±1000)	9.0 (±3)	6.0 (±2)	5.0 (±2)	1.0 (±2)	<1
48	'Lapin'	12	57.1	200 (±100)	32 (±6)	6.0 (±1)	4.0 (±0)	3.0 (±0)	3.0 (±0)
48	'Rainier'	12	53.1	4500 (±700)	1900 (±300)	160 (±100)	1.0 (±1)	<1	<1
48	'Sweetheart'	12	51.2	830 (±300)	59 (±30)	11 (±1)	¶[b]	<1	<1
48	'Tulare'	12	55.4	6500 (±2000)	140 (±100)	8.0 (±2)	5.0 (±1)	1.0 (±1)	<1
64	'Brooks'	6	54.9	7300 (±800)	13 (±3)	7.0 (±1)	5.0 (±1)	<1	<1
64	'Garnet'	6	58.8	22 000 (±1000)	5000 (±2000)	400 (±300)	120 (±80)	<1	<1
64	'Lapin'	6	60.1	400 (±300)	50 (±20)	10 (±3)	5.0 (±0)	4.0 (±0)	3.0 (±0)
64	'Rainier'	6	59.2	16 000 (±700)	5400 (±0)	1300 (±300)	410 (±200)	<1	<1
64	'Sweetheart'	6	47.1	1500 (±300)	160 (±70)	19 (±2)	3.0 (±0)	<1	<1
64	'Tulare'	6	55.5	10 000 (±2000)	34 (±5)	8.0 (±3)	4.0 (±1)	1.0 (±1)	<1

[a] $n = 3$.
[b] ¶ Missing data.

<1 μg kg^{-1} after three days (for 'May Grand').[8] Tolerances for methyl bromide have not been established.

Bromide ion residues among the cultivars were not statistically different (Table 2). 'Fuji' had the lowest concentration (3.3 (±0.3) mg kg^{-1}) whereas 'Jonagold' had the highest concentration (4.9 (±0.6) mg kg^{-1}). Singh *et al*[6] observed <4 mg kg^{-1} after three weeks aeration at 0 °C in 'Granny Smith' apples fumigated in fibreboard cartons with 32 g m^{-3} for 2 h at 7 °C. The tolerance level of bromide ion in apples is 5 mg kg^{-1} for the United States and 20 mg kg^{-1} for Australia.[6,18]

3.2 Sweet cherries

Methyl bromide residues declined rapidly after a 2 h aeration and 5 h holding period (Table 3). The first observations for the 'Lapin' and 'Sweetheart' cherries may be lower than the other cultivars because of the improved ventilation system during aeration at the newer laboratory where they were fumigated. Only 'Lapin' had methyl bromide residues >2 μg kg^{-1} five days after fumigation. The highest and longest-lasting residues were with the 64 g m^{-3} treatments. Using 'Bing' cultivars, Moffitt *et al*[19] found <0.42 mg kg^{-1} methyl bromide after methyl bromide fumigation with 64 g m^{-3} for 2 h at 6.5 °C and Tebbets *et al*[7] recorded <0.19 mg kg^{-1} a day after methyl bromide fumigation with 48 g m^{-3} for 2 h at 21 °C.

The bromide ion residues were consistent among the cherry cultivars and were slightly higher than those for the apples (Table 2). Moffitt *et al*[19] observed ≤6.93 mg kg^{-1} of bromide ion after methyl bromide fumigation with 48 g m^{-3} for 2 h at ≃12 °C. Singh *et al*[6] evaluated a range of cultivars ('Early Lyons,' 'Eagle Seedling,' 'St. Margaret,' 'Rons Seedling,' and 'Lambert') and reported <8 mg kg^{-1} after methyl bromide fumigation with 48 g m^{-3} for 2 h at 15 °C. The EPA tolerance and Japanese maximum residue limit for bromide ion in sweet cherries is 20 mg kg^{-1}.[18,20]

3.3 Desorption rates

Hartsell *et al.*[13] found that methyl bromide residues in fumigated nectarines declined with time following a power curve equation. Tebbets *et al.*[7] worked with a variety of fumigated fruits and developed a logarithmic equation similar to

$$\ln c = \ln c_0 - kt \tag{2}$$

Table 4. Parameters of a descriptive model (eqn. (2)) of methyl bromide desorption from regression analysis. Intercept $\ln c_0$ is extrapolated concentration at beginning of desorption ($t = 0$).

				Regression coefficient		*Intercept*		
Fruit	*Cultivar*	*Dose (g m^{-3})*	*r^2*	*k*	*SE*	*ln c_0*	*SE*	*Half-life[a] (days)*
Apple	'Braeburn'	56	0.993	0.53	0.02	8.8	0.1	1.31
	'Gala'	56	0.923	0.75	0.09	8.5	0.7	0.92
	'Fuji'	56	0.997	0.71	0.02	9	0.1	0.98
	'Granny Smith'	56	0.988	0.67	0.03	8.8	0.3	1.03
	'Jonagold'	56	0.954	0.8	0.07	8.3	0.5	0.87
Cherry	'Brooks'	32	0.685	0.9	0.3	4	1	0.81
		40	0.701	0.9	0.3	4	1	0.78
		48	0.658	1	0.4	5	1	0.68
		64	0.666	1.1	0.4	6	2	0.62
Cherry	'Garnet'	32	0.602	1.4	0.6	3	2	0.5
		40	0.875	1.8	0.4	6	1	0.38
		48	0.911	1.8	0.3	7	1	0.39
		64	0.994	2.2	0.1	10	0	0.32
	'Lapin'	32	0.846	0.4	0.1	2.7	0.3	1.68
		40	0.67	0.3	0.1	2.6	0.5	2.03
		48	0.667	0.5	0.2	3.9	0.7	1.36
		64	0.713	0.6	0.2	4.5	0.7	1.14
	'Rainier'	32	0.612	1.4	0.5	3	2	0.51
		40	0.646	1	0.4	1	2	0.7
		48	0.939	1.9	0.2	8	1	0.37
		64	0.972	2.2	0.2	11	1	0.32
Cherry	'Sweetheart'	32	0.7	0.8	0.3	4	1	0.89
		40	0.723	0.9	0.3	4	1	0.79
		48	0.872	1	0.2	5.5	1	0.67
		64	0.855	1.2	0.2	6	1	0.6
	'Tulare'	32	0.686	0.8	0.3	4	1	0.83
		40	0.692	0.9	0.3	5	1	0.76
		48	0.784	1.2	0.3	6	1	0.57
		64	0.703	1.2	0.4	6	1	0.59

[a] Half life = ln 2/*k*.

to describe residue reduction over time; the correlation coefficients of this descriptive model were high for plums, pears, cherries nectarines, and peaches ($r > 0.955$). In our studies, this model (eqn (2)) also had high correlations ($r > 0.970$) regardless of load factor (the ratio of packed fruit volume to chamber volume) and pulp temperature in cherries and in apples (Table 4).[9,10]

The parameter k may be considered a *desorption rate constant* (h^{-1}). The larger the value of k, the more quickly the residue dissipates. Large k values are associated with increased pulp temperature, particularly during the first 24 h after aeration.[9,10] In our studies with cherry cultivars, the value of k tended to increase with initial dose (Table 4). The temperature effect may not be evident here because the samples were frozen after fumigation and some cell disruption may have occurred on thawing which may have altered desorption rate. Tebbets *et al.*[7] found the greatest value of k with pears and plum, then cherries, and finally peaches and nectarines. If k is divided into the natural logarithm of 2, the result is the half-life of the residues. In our studies, the half-lives of the residues were very short for all cultivars (Table 4). Although residue desorption data from cultivars of both commodities were described by the logarithmic model (eqn (2), the apple cultivars generally had the better fit (Table 4). None of the apple and cherry model parameters was significantly different among the cultivars for each fumigation dose except between 'Garnet' and 'Lapin' for $64\,g\,m^{-3}$ ($t = 2.98$, df = 2, $P < 0.05$). Desorption models are important because they can accurately describe the persistence of methyl bromide with duration of aeration.[9,10] The fruits, regardless of cultivar, need to have sufficient time to dissipate methyl bromide before storage in a confined space. Commercial exporters can use the desorption models to schedule adequate aeration time and to obtain allowable residues.

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